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DELIVERABLE (D-N<sup>o</sup>: 2.1)

# Status of fission gas release studies (34 months)

FIRST-Nuclides

(Contract Number: FP7-295722)

D.H. Wegen, O. Beneš, E. González-Robles, A.  
Puranen

2014



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Classification: No restriction  
Unit: E05 and E02  
Action No:51102

European Commission  
Joint Research Centre  
Institute for Transuranium Elements

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Author(s): D.H. Wegen, O. Beneš, E. González-Robles, A. Puranen

Reporting period: 01/01/14 – 30/09/14

Date of issue of this report: 31/10/14

Start date of project: 02/01/12

Duration: 36 Months

<b>Project co-funded by the European Commission under the Seventh Euratom Framework Programme for Nuclear Research &amp; Training Activities (2007-2011)</b>		
<b>Dissemination Level</b>		
<b>PU</b>	Public	X
<b>RE</b>	Restricted to a group specified by the partners of the <a href="#">FIRST-Nuclides</a> project	
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## Status of fission gas release studies (34 months)

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### Objectives

In the first component of work package 2 (WP2) “Experimental determination of fission gas release” the focus is on the quantification of fission gases and fission gas release in high burn-up (HBU) UO<sub>2</sub> spent nuclear fuels (SNF). Fission gas sampled in the plenum of a fuel rod are analysed as well as the grain boundary inventory and the cross sectional distribution of fission gases and volatile fission products. The experimental part in WP2 started in project month 4 and ended in project month 33 [1], [4], [10], [2].

The JOINT RESEARCH CENTRE – INSTITUTE FOR TRANSURANIUM ELEMENTS (JRC-ITU) is the leading organization of WP2. In the first project year the fission gas release (FGR) from a spent fuel rod owned by KIT was measured [1], [3]. The determination of the inventory of fission gas and fission products in grain boundaries were foreseen for the third project year.

The KARLSRUHER INSTITUT FÜR TECHNOLOGIE (KIT) analysed in the first project year fission and activation products in the fission gas sampled at JRC-ITU from the plenum of a fuel rod segment by puncturing. The development, testing and implementation of analytical methods for fission and activation products have started in the first project year one and were continued in the second. Leaching experiments in which gas and solution analyses are foreseen were started in the first year and last until project month 33 [1].

STUDSVIK NUCLEAR AB (STUDSVIK) investigates in the frame of WP2, the radial fission gas and volatile fission product distribution (Xe, I, and Cs) by Laser-Ablation Mass Spectroscopy (LA-MS) on HBU boiling water reactor (BWR) SNF [1].

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## Status and results

KIT continued the leaching experiment under anoxic conditions which was started in the 2<sup>nd</sup> project year. A clad segment pellet was introduced in an autoclave together with 220 ml of bicarbonate water (19 mM NaHCO<sub>3</sub> + 1 mM NaCl). Then the autoclave was pressurised with Ar/H<sub>2</sub> (p<sub>T</sub> = 40 bar; p<sub>H<sub>2</sub></sub> = 3 bar). First gas samples have been analysed. After a cumulative contact time of 57 days, 4.3% of the Xe and 17% of the Kr inventories were released into the gas phase [5], [6], [11], [12], [13], [16], [17].

STUDSVIK finalized the analysis of Laser Ablation data obtained on cross sections from a standard UO<sub>2</sub> fuel and an Al/Cr-additive fuel. The findings of the Laser Ablation study on both pellets indicate cesium and iodine profiles that are very similar and appear to follow the radial burn up profile (as indicated by <sup>140</sup>Ce). Cesium, iodine and to some extent selenium also appear to collect in some fuel cracks. Selenium was tentatively identified by the good agreement of the isotopic ratios of mass 77, 79 and 82 with the calculated inventory. For the additive pellet chromium and especially aluminum are heterogeneously distributed in the pellet. Details are given in [7], [8], [14], [15], [16].

JRC-ITU has determined the inventory of fission gas and fission products in grain boundaries by Knudsen cell effusion tests in the coming project year.

JRC-ITU has examined a fine powder of ~5mg quantity of irradiated BWR UO<sub>2</sub> with average burn-up 54Gwd/t<sub>HM</sub> which was drilled from the core region of the fuel pin. The sample was put in the Knudsen cell and was heated under vacuum conditions at rate of 10 K/min until complete vaporization of the fuel at 2460 K. During the measurement the atomic mass units corresponding to relevant isotopes of examined fission products have been recorded. Indications for two release mechanisms for <sup>137</sup>Cs, one with rather low intensity starting just below 1000 K and a significant release after 1500 K have been detected. The release of the examined fission products <sup>88</sup>Sr, <sup>87</sup>Rb, <sup>137</sup>Cs, and <sup>136</sup>Xe has been semi-quantified [9].

## Dissemination

Publications, reports, or contributions in reports, proceedings:

- [1] Wegen, D.H., González-Robles, E., Puranen, A. (2012). *DELIVERABLE (D-N°: 2.1) - Status of fission gas release studies (12 months), FIRST Nuclides (Contract Number: FP7-295722)*. JRC Scientific and Policy Reports, JRC76116, European Atomic Energy Community, Germany.
- [2] Wegen, D.H., González-Robles, E., Puranen, A. (2013). *DELIVERABLE (D-N°: 2.1) - Status of fission gas release studies (24 months), FIRST Nuclides (Contract Number: FP7-295722)*. JRC Scientific and Policy Reports, JRC87028, European Atomic Energy Community, Germany.
- [3] Wegen, D.H., Papaioannou, D., De Weerd, W., Rondinella, V.V., Glatz, J.-P. (2013). *Fission gas release measurement on one 50.4 GWD/tHM PWR fuel segment*. 1<sup>st</sup> Annual Workshop

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Proceedings, 7th EC FP – FIRST-Nuclides, 9<sup>th</sup> – 11<sup>th</sup> October 2012, Budapest, Hungary. KIT SCIENTIFIC REPORTS 7639, pp 201.

- [4] Wegen, D.H. (2013). *Overview WP2: Gas Release + RIM and Grain Boundary Diffusion*. Proceedings of the 2<sup>nd</sup> Annual Workshop of the FIRST- Nuclides Project, November 5<sup>th</sup>-7<sup>th</sup>, Antwerp, Belgium.
- [5] González-Robles. E., Bohnert, E., Loida, A., Müller, N., Metz, V., Kienzler, B. (2013). *Fission gas measurements and description of leaching experiments with of KIT's irradiated PWR fuel rod segment (50.4 GWd/t<sub>HM</sub>)*. 1<sup>st</sup> Annual Workshop Proceedings of the Collaborative Project “Fast /Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel” (7<sup>th</sup> EC FP CPFIRST-Nuclides), 9-11 October 2012, Budapest (Hungary), KIT SCIENTIFIC REPORTS 7639, pp 231.
- [6] González-Robles, E., Bohnert, E., Müller, N, Herm, M., Metz, V. (2013). *Determination of the fission gas release in the segment N0204 and gas phase result of anoxic leaching experiment*. Proceedings of the 2<sup>nd</sup> Annual Workshop of the FIRST- Nuclides Project, November 5<sup>th</sup>-7<sup>th</sup>, Antwerp, Belgium.
- [7] Roth, O., Puranen, A., Low, J., Granfors, M, Cui, D., Askeljung, C. (2013). *Spent fuel leaching experiments and laser ablation studies performed in Studsvik - Status and preliminary results*. Proceedings of the 2<sup>nd</sup> Annual Workshop of the FIRST- Nuclides Project, November 5<sup>th</sup>-7<sup>th</sup>, Antwerp, Belgium.
- [8] Puranen, A., Granfors, M., Roth, O., (2014). *Laser ablation study of irradiated standard UO<sub>2</sub> fuel and Al/Cr doped UO<sub>2</sub> fuel*. Proceedings of the Final Annual Workshop of the FIRST- Nuclides Project, September 1<sup>st</sup>-2<sup>nd</sup>, Karlsruhe, Germany.
- [9] Colle, J.-Y., Beneš, O., Serrano-Purroy D., Sureda Pastor, R., Martinez Torrents, A. (2014). *Fission product release from irradiated fuel*. Proceedings of the Final Annual Workshop of the FIRST- Nuclides Project, September 1<sup>st</sup>-2<sup>nd</sup>, Karlsruhe, Germany.

#### Poster and presentations:

- [10] Wegen, D.H. (2013). *WP2: Gas Release & Rim and Grain Boundary Diffusion*. 2<sup>nd</sup> Annual Workshop, 7<sup>th</sup> EC FP – FIRST-Nuclides, 5<sup>th</sup>-7<sup>th</sup> November, Antwerp, Belgium.
- [11] González-Robles, E., Wegen, D.H., Papaioannou, D., Kienzler, B., Nasyrow, R., Metz, V. (2013). *Physical characterisation of spent nuclear fuel: First steps to further Instant Release Fractions investigations*. 8<sup>th</sup> EC Conference on the Management of Radioactive Waste, EURADWASTE 2013, October 14<sup>th</sup>-17<sup>th</sup>, Vilnius, Lithuania.
- [12] González-Robles, E., Bohnert, E., Metz, V., Wegen, D.H., Papaioannou, D., Kienzler, B. (2013). *Physical characterisation and calculation of the initial and boundary conditions of a commercial UO<sub>2</sub> spent nuclear fuel regarding the radionuclide release*. 37<sup>th</sup> Symposium on the Scientific Basis for Nuclear Waste Management, September 29<sup>th</sup>–October 3<sup>rd</sup>, Barcelona, Spain.

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- [13] González-Robles, E., Bohnert, E., Müller, N, Herm, M., Metz, V. (2013). *Determination of the fission gas release in the segment N0204 and gas phase result of anoxic leaching experiment*. 2<sup>nd</sup> Annual Workshop of the FIRST- Nuclides Project, November 5<sup>th</sup>-7<sup>th</sup>, Antwerp, Belgium.
- [14] Puranen, A., Roth, O., Granfors, M. (2013). *Investigating the radial distribution of potential rapid release radionuclides in irradiated nuclear fuel*. Symposium: E No. 2 15, E-MRS 2013 Spring Meeting, May 27-31, Strasbourg, France.
- [15] Puranen, A., Granfors, M, Roth, O. (2013). *Laser ablation experiments at Studsvik*. 2<sup>nd</sup> Annual Workshop of the FIRST- Nuclides Project, November 5<sup>th</sup>-7<sup>th</sup>, Antwerp, Belgium.
- [16] Wegen, D.H. (2014). *WP2 Summary: Gas Release & Rim and Grain Boundary Diffusion*. 3<sup>rd</sup> Annual Workshop, 7<sup>th</sup> EC FP – FIRST-Nuclides, September 1<sup>st</sup>-2<sup>nd</sup>, Karlsruhe, Germany.
- [17] González-Robles, E. (2014). *Fast radionuclide release from a PWR fuel rod segment (50.4 GWd/tHM) under hydrogen overpressure*. Spent Fuel Workshop 2014, September 3<sup>rd</sup>-5<sup>th</sup>, Karlsruhe, Germany.

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Author(s): D.H. Wegen, O. Beneš, E. González-Robles, A. Puranen

**Abstract:** This report summarises the activities planned and performed in project months 25 - 33 by the beneficiaries collaborating in the component “*Experimental determination of fission gas release*” of work package 2 (WP2) of the CP – FIRST-Nuclides project in 2014. The main achievements in the final project year are given.

The research leading to these results has received funding from the European Union’s European Atomic Energy Community’s (EURATOM) Seventh Framework Programme FP7/2007-2011 under grant agreement no. 295722 (FIRST-Nuclides project).

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